

The present invention relates to a process for oligomerizing  $C_6$ -olefins, in particular for preparing  $C_{12}$ -olefins by dimerization.

As described in the specification beginning at page 1, line 8, processes for the oligomerization of olefins are known, such as that disclosed in DE-A-4339713, which is the German equivalent of Vicari et al. As described in the specification herein at page 1, lines 9-14, Vicari et al. describes a process for oligomerizing olefins to give highly linear oligomers. In this process,  $C_{2-6}$ -olefins are reacted at superatmospheric pressure and elevated temperature over a fixed-bed catalyst comprising from 10 to 70% by weight of nickel oxide, from 5 to 30% by weight of titanium dioxide and/or zirconium dioxide, from 0 to 20% by weight of aluminum oxide as significant active constituents and silicon dioxide as the remainder.

As described in the specification at page 1, line 25, a disadvantage of the known processes is that the catalyst life is frequently too short. The catalyst is, in particular, clogged by higher oligomers and therefore loses its activity.

Applicants have discovered that by oligomerizing  $C_6$ -olefins using the above-discussed fixed-bed catalyst of Vicari et al. at a conversion to oligomerized  $C_6$ -olefins of not more than 30% by weight, based on the reaction mixture, deactivation of the catalyst can be avoided and dimer selectivity can be increased. By recycling unreacted reaction mixture after separating off the oligomers, a higher total conversion can be obtained. Indeed, the presently-claimed process makes it possible to realize a total conversion of over 90% together with a  $C_{12}$  (dimer) selectivity of over 80%.

Adherence to the conversion specified according to the claims greatly increases the operating life of the catalyst, since the formation of high-boiling compounds which can

deposit on the catalyst and thus cause a drop in activity is suppressed, as described in the specification at page 2, lines 22-27.

During the above-referenced interview, the Examiner commented that there is no data in the specification demonstrating increased catalyst operating life. Applicants' reply is that since the Examiner has provided no evidence to suggest that catalyst operating life is not improved by the present invention, Applicants' have no burden to disprove such evidence. Moreover, Applicants, their attorneys, and all associated with the filing and prosecution of the application are under a duty of candor and good faith under 37 CFR 1.56. The Examiner has no reason to question the truth of Applicant's assertion regarding catalyst life.

While Vicari et al discloses the presently-recited fixed-bed catalyst, as discussed above, Vicari et al do not recognize the nexus between maximum conversion, on the one hand, and increased catalyst life, higher selectivity of dimer, and higher overall conversion, on the other hand.

Examples 1 and 2 illustrate the presently-claimed invention, and the above-discussed superior results. In Example 1, beginning with a hydrocarbon mixture feed containing 73% by weight of  $C_6$  component, a  $C_6$ -olefin conversion of 94.7% and a  $C_{12}$  selectivity of 83.6%, based on the  $C_6$ -olefins reacted, was obtained.

In Example 2, the  $C_6$  concentration in the hydrocarbon mixture feed was 98.7% by weight. The result was a  $C_6$ -olefin conversion of 98.4% and a  $C_{12}$  selectivity of 82.6%, based on the  $C_6$ -olefins reacted.

During the above-referenced interview, the Examiner queried how the results in Examples 1 and 2 square with the above-discussed limitation of a maximum conversion of not more than 30% by weight. Applicants' attorney and the assignee's representative explained, referring to Figs. 1A or 1B, that the 30% conversion is with respect to the input to,

and output from, reactor C1, while the final conversion and selectivity of the Examples is with respect to bottoms stream S. The conversion regarding input to and output from C1 for each Example was calculated and is 20.2% for Example 1, and 19.4% for Example 2, as disclosed in the specification in the "Total C<sub>9</sub>-" column and "Feed mixture to K1=reactor output" row, at the top of page 9, and the bottom of page 10, respectively.

As discussed above, Vicari et al do not recognize any nexus between conversion and the other listed properties. Indeed, based on the examples and comparative examples in Vicari et al, the conversion varies from 41-55% for the comparative examples, and 53-78% for the examples. See the various tables at column 7 therein. Since no recycle is disclosed, it is assumed that these conversions are ultimate conversions. While this data is for C<sub>4</sub>-olefins, nevertheless, it is still sufficient to show that Vicari et al do not recognize the significance of maintaining the conversion below a certain maximum for each single pass over the catalyst.

Sanderson et al does not remedy any of the above-discussed deficiencies of Vicari et al. Sanderson et al discloses a process for oligomerizing olefins using sulfate-activated molecular sieves. Sanderson disclose that by the use of such molecular sieves, a high percentage of dimers, i.e., a high dimer to trimer ratio, can be achieved (column 2, lines 35-37). Sanderson et al further disclose that the oligomerization results in an oligomer product distribution that varies with reaction time; as the reaction time increases, the olefin monomer conversion increases, and the selectivities for the heavier oligomers increase (column 3, lines 33-55). The olefin monomer feed stocks of Sanderson et al must contain at least 10 carbon atoms (column 3, lines 13-21).

In Sanderson et al, no particular olefin conversion goal is disclosed, although it is noted that liquid hourly space velocity may be varied within wide limits (column 5, lines 55-61), and it is noted that the olefin conversion, as specified in the table bridging columns 7 and

8, varies widely, from 3.0% to 70.4%. Moreover, the data therein show that conversion is not strictly a function of reaction time, as can be seen by comparing examples using the same catalyst. For example, Catalyst 4 produces a conversion of 3% in 5 hours (Example 1), and a conversion of 11.3% in 4 hours, but at a higher temperature (Example 7). A comparison of Examples 2 and 6; Examples 4 and 5; produces a similar result. Thus, the conversion was actually lower after a longer reaction time. In addition, all the examples therein employed a reaction time of 4 or 5 hours, yet no trend can be discerned therefrom that conversion must be limited to 30% or lower to maximize dimer product. Rather, the trend is due to the use of sulfate-activated molecular sieves (Catalysts 2-4 and 7) compared to the non-sulfate-activated molecular sieves (Catalysts 1, 5 and 6). Compare Examples 3 and 5, where at identical feeds, reaction times and temperatures, the sulfated molecular sieve produced a conversion of 32.5% (Catalyst 2), while the same catalyst in non-sulfated form produced a conversion of 18.8% (Catalyst 1). Compare also Examples 11 and 13, which also shows increased conversion using the sulfated molecular sieve.

Sanderson et al adds nothing to Vicari et al. Sanderson et al is concerned with a different catalyst therefrom, and olefins having a carbon range outside the range specified by Vicari et al. Sanderson et al disclose nothing with regard to any possibility of increased catalyst life by limiting conversion percentage to a particular amount. Nor would one skilled in the art even combine Vicari et al and Sanderson et al in view of the mutually exclusive catalysts and olefins used. Indeed, without the present disclosure as a guide, one skilled in the art would not be motivated to combine Vicari et al and Sanderson et al. Even if combined, the result would not be the presently-claimed invention.

During the above-referenced interview, the Examiner took the position, erroneously in Applicants' view, that a demonstration of increase catalyst life was necessary to establish

patentability over the applied combination of prior art. While Applicants submit that they have shown patentability in the absence of such demonstration, for reasons discussed above, nevertheless, the newly-submitted Heidemann Declaration provides such a demonstration.

For all the above reasons, it is respectfully requested that the rejection over Vicari et al in view of Sanderson et al be withdrawn.

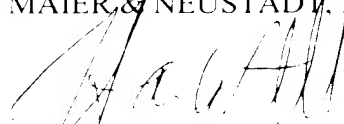
The rejection of Claim 9 under 35 U.S.C. § 112, second paragraph, is respectfully traversed. The function of the protective bed is defined in the specification, and is now recited in the claims. In addition, Figs. 1A and 1B demonstrate that the protective bed, designated as "F1" can be separate from the reactor and the column. During the above-referenced interview, the Examiner indicated that the above amendment would overcome this rejection. Accordingly, it is respectfully requested that this rejection be withdrawn.

All of the presently pending claims in this application are now believed to be in

immediate condition for allowance. Accordingly, the Examiner is respectfully requested to pass this application to issue.

Respectfully submitted,

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Amendment Filed on  
Herewith:

IN THE SPECIFICATION

Replace the paragraph at page 8, lines 7-8 as follows:

--The hydrocarbon mixture was introduced into the column K1 (Fig. 1A) at a rate of 5.1 kg/h. The following conditions were set in the experimental plant:--

Replace the paragraph at page 9, last two lines as follows:

--The hydrocarbon mixture was introduced into the filter F1 (Fig. [2] 1B) at a rate of 3.20 kg/h. The following conditions were set in the experimental plant:--

IN THE CLAIMS

--9. (Twice Amended) A process as claimed in claim 1, wherein the reaction is passed over a protective bed wherein catalyst poisons are removed, prior to the reaction.--